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(54) Separator for fuel cell, process for production thereof, and solid polymer type fuel cell using said separator

(57) A separator for fuel cell, formed using a base material obtained from a composition comprising at least a binder, a powdery carbon filler having an average particle diameter of 10 nm to 100 µm, and a short fiber having an average fiber length of 0.03 to 6 mm, in which composition the amount ratio of the above three components is such that the amount of the powdery carbon filler is 200 to 800 parts by weight and the amount of the short fiber is 10 to 300 parts by weight, both per 100 parts by weight of the binder.

The above fuel cell separator alleviates the problems of the prior art, has a strength necessary for separator even when made in a small thickness, generates no crack at the time of fuel cell assembling, and has high reliability.

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Description**Background of the Invention**5 **(1) Field of the Invention**

[0001] The present invention relates to a separator for fuel cell, a process for production thereof, and a solid polymer type fuel cell using the separator. More particularly, the present invention relates to a separator for fuel cell having a strength necessary for separator even when made in a small thickness, a process for production thereof, and a solid polymer type fuel cell using the separator.

(2) Description of the Prior Art

[0002] Fuel cells are advantageous in that they require substantially no fossil fuel (whose depletion in the near future is feared) and, when used for power generation, generate substantially no noise and are high in energy recovery as compared with other methods for power generation. Therefore, fuel cells are being developed as a power generation plant of relatively small size for buildings and factories.

[0003] Among fuel cells, a solid polymer type fuel cell operates at lower temperatures than other types of fuel cell do; therefore, it has such characteristics that not only the parts constituting the cell are little corroded, but also the cell can discharge a relatively large electric current for the low temperature operation. Therefore, the solid polymer type fuel cell is drawing attention as a substitute electric source for internal combustion engine of automobile.

[0004] Currently, a smaller size or thickness is required for various products and parts. In the solid polymer type fuel cell as well, a small volume and a compact shape are required when it is used as a substitute electric source for internal combustion engine of automobile.

[0005] Among the parts constituting the solid polymer type fuel cell, the separator is generally a flat plate having a plurality of parallel grooves formed at one or both sides, and is used for transferring the electricity generated inside the fuel cell, to outside.

[0006] Some of the conventional fuel cell separators are produced by impregnating a high-density graphite or a graphite material with a thermosetting resin and forming, at one or both sides of the resulting material, a plurality of parallel grooves by machining.

[0007] In JP-B-1-57466 is proposed a process for producing a separator by subjecting a mixture of a particular thermosetting resin and a graphite powder to pressure molding.

[0008] In JP-A-4-214072 is proposed a process for producing a fuel cell separator by molding a mixture of a phenolic resin, a milled carbon fiber and a graphite powder into a sheet shape and graphitizing the sheet material.

[0009] As a fuel cell of small volume and compact shape has come to be required as mentioned previously, the separator of fuel cell (which occupies a large portion of fuel cell) inevitably needs to be formable in a small size or in a small thickness and, when formed in a small thickness, needs to have a strength required for separator; that is, the separator needs to have a high strength and high toughness. However, the conventional separator obtained by subjecting graphite to machining and the separator obtained by subjecting, to pressure molding, a mixture of a particular thermosetting resin (specifically, a very special phenolic resin having a p-xylene bond in the molecular chain) and a graphite powder have had problems in that these separators, when made in a small thickness, are fragile, easily break at the time of fuel cell assembling, and have low reliability.

[0010] The fuel cell separator of small thickness obtained by subjecting a mixture of a resin, a fiber and a graphite powder to graphitization, has had problems in that (1) since the graphitization need be conducted by firing a sheet-shaped material at a high temperature of 1,500 to 3,000°C, the resulting separator is expensive owing to the use of a special equipment and method for such graphitization, (2) the sheet-shaped material contracts during the graphitization, resulting in low dimensional reproducibility, and (3) the strain caused by the above contraction remains in the graphitization product.

50 **Summary of the Invention**

[0011] The present invention has been made with an aim of providing a separator for fuel cell which alleviates the problems of the prior art, has a strength necessary for separator even when made in a small thickness, generates no crack at the time of fuel cell assembling, and has high reliability; a process for producing the above separator for fuel cell, which requires no graphitization step; and a solid polymer type fuel cell of excellent durability using the above separator.

[0012] According to the present invention, there is provided a separator for fuel cell, formed using a base material obtained from a composition comprising at least a binder, a powdery carbon filler having an average particle diameter

of 10 nm to 100 μm , and a short fiber having an average fiber length of 0.03 to 6 mm, in which composition the amount ratio of the above three components is such that the amount of the powdery carbon filler is 200 to 800 parts by weight and the amount of the short fiber is 10 to 300 parts by weight, both per 100 parts by weight of the binder.

[0013] The present invention further provides a process for producing a separator for fuel cell, which comprises

5 mixing at least a binder, a powdery carbon filler having an average particle diameter of 10 nm to 100 μm , and a short fiber having an average fiber length of 0.03 to 6 mm in such an amount ratio that the amount of the powdery carbon filler becomes 200 to 800 parts by weight and the amount of the short fiber becomes 10 to 300 parts by weight, both per 100 parts by weight of the binder, granulating the resulting mixture into a granular material of 0.03 to 5 mm in particle diameter, and molding the granular material into a separator shape.

10 [0014] The present invention furthermore provides a solid polymer type fuel cell using the above separator.

Detailed Description of the Invention

[0015] The present invention is described in detail below.

15 [0016] The binder used in the present invention has no particular restriction as long as it is a material which is so called. There can be mentioned, for example, a thermosetting resin, a thermoplastic resin and a rubber.

[0017] As the thermosetting resin, there can be mentioned, for example, phenolic resin, polycarbodiimide resin, furfuryl alcohol resin, epoxy resin, cellulose, urea resin, melamine resin, unsaturated polyester resin, silicone resin, diallyl phthalate resin, bismaleimidetriazine resin, polyaminobismaleimide resin and aromatic polyimide resin. They can be 20 used singly or in admixture of two or more kinds.

[0018] As the thermoplastic resin, there can be mentioned, for example, polyethylene, polystyrene, polypropylene, polymethyl methacrylate, polyethylene terephthalate, polybutylene terephthalate, polyethersulfone, polycarbonate, polyoxamethylene, polyamide, polyimide, polyamideimide, polyvinyl alcohol, polyvinyl chloride, polyphenylsulfone, polycarbonate ether ketone, polysulfone, polyether ketone, polyarylate, polyetherimide, polymethylpentene, fluororesin, polyoxybenzoyl ester resin, liquid crystal polyester resin, aromatic polyester, polyacetal, polyallylsulfone, polybenzimidazole, polyethernitrile, polythioethersulfone and polyphenylene ether. They can be used singly or in admixture of two or more kinds.

[0019] As the rubber, there can be mentioned, for example, fluororubber, silicone rubber, butyl rubber, chloroprene rubber, nitrile rubber, nitrile-chloroprene rubber, chlorinated butyl rubber, epichlorohydrin rubber, epichlorohydrin-ethylene oxide rubber, epichlorohydrin-ethylene oxide-acrylic glycidyl ether terpolymer, urethane rubber, acrylic rubber, ethylene-propylene rubber, styrene rubber, butadiene rubber and natural rubber. They can be used singly or in admixture of two or more kinds.

[0020] The powdery carbon filler used in the present invention has no particular restriction except the particle diameter explained later, as long as it is a powdery carbon filler having excellent electroconductivity. There can be mentioned, for example, those made of natural graphite (e.g. scaly graphite or lumpy graphite), expanded graphite, artificial graphite, mesophase carbon, coal coke, petroleum coke, acetylene black, carbon black, Ketjen Black and glassy carbon.

[0021] The particle diameter of the powdery carbon filler can be 10 nm to 100 μm , preferably 20 to 80 μm in terms of average particle diameter. When the average particle diameter of the powdery carbon filler is smaller than 10 nm, bulk density of the powdery carbon filler is too low, which may allow the resulting separator to have a high electrical resistance or may make difficult the homogeneous mixing of the powdery carbon filler with the binder component (heterogeneous mixing may incur inferior molding). When the average particle diameter is larger than 100 μm , electrical contact is low between the particles of the powdery carbon filler or between the powdery carbon filler and the short fiber, which may make high the electrical resistance of the resulting separator.

[0022] The short fiber used in the present invention is preferably one made from a material high in strength and modulus of elasticity and excellent in corrosion resistance, electroconductivity, heat resistance and electrochemical corrosion resistance. There can be used, for example, at least one kind selected from a carbon fiber, a pyrolyzed carbon fiber (a flame-retarded fiber) and a stainless steel fiber.

[0023] The fiber length of the short fiber can be 0.03 to 6 mm, preferably 0.07 to 3.0 mm, more preferably 0.1 to 0.9 mm in terms of average fiber length. When the average fiber length of the short fiber is smaller than 0.03 mm, no strength improvement is obtained and, when a separator of small thickness is produced with this short fiber, the separator has no sufficient strength and may be unusable as a separator for fuel cell. When the average fiber length of the short fiber is larger than 6 mm, entanglement between fibers takes place in the mixing step of components, resulting in no sufficient dispersion of the short fiber; as a result, heterogeneity of strength arises and cracking may appear at the time of fuel cell assembling or during the operation of fuel cell.

[0024] As the carbon fiber, there can be mentioned a fiber obtained by subjecting a raw material (e.g. pitch-based fiber, polyacrylonitrile fiber, phenolic fiber, rayon fiber, cellulose-based fiber or aramid fiber) to a heat treatment of 450 to 3,000 °C, preferably 800 to 2,200°C in an inert gas atmosphere. As the pyrolyzed carbon fiber, there can be men-

tioned a fiber obtained by heating the same material as used for the carbon fiber, at a temperature of about 200 to 450°C.

[0025] To the mixture of the binder, the powdery carbon filler and the short fiber may be added a lubricant in order to improve the releasability from die during molding. There is no particular restriction as to the kind of the lubricant used, but there can be mentioned, for example, a fatty acid or a metal salt thereof (a metal soap), such as stearic acid, zinc stearate or the like; a fatty acid ester; a fatty acid chloride; an amine type fatty acid; an amide type fatty acid; a wax; and a polymer wax.

[0026] The amount ratio of components in the mixture of the binder, the powdery carbon filler and the short fiber can be such that the amount of the powdery carbon filler is 200 to 800 parts by weight and the amount of the short fiber is 10 to 300 parts by weight, both per 100 parts by weight of the binder.

[0027] When the amount of the powdery carbon filler is smaller than 200 parts by weight, the resulting separator has insufficient electroconductivity, particularly insufficient conductive paths between fibers and may have a high resistance. When the amount is larger than 800 parts by weight, the separator has an insufficient strength and, when made in a small thickness, generates cracks at the time of fuel cell assembling or during power generation and may be unusable as a separator for fuel cell.

[0028] When the amount of the short fiber is smaller than 10 parts by weight, the resulting separator has an insufficient strength and, when made in a small thickness, generates cracks at the time of fuel cell assembling or during power generation. When the amount is larger than 300 parts by weight, molding becomes inferior. In both cases, the separator obtained may be unusable as a separator for fuel cell.

[0029] The fuel cell separator of the present invention is produced according to the process of the present invention. First, the binder, the powdery carbon filler and the short fiber are mixed in the above-mentioned amount ratio; the resulting mixture is as necessary mixed with a solvent and subjected to granulation. This mixing and granulation step can be conducted by a known mixing method using, for example, a stirring rod, a kneader, a mixer, a static mixer, a Henschel mixer, a super mixer or a ribbon mixer. There is no particular restriction as to the mixing method.

[0030] As the solvent used as necessary in the mixing and granulation step, there can be mentioned, for example, water, methanol, ethanol, isopropyl alcohol, butanol, toluene, xylene, methyl ethyl ketone and acetone. However, the solvent is not restricted to these. The amount of the solvent used is 40% by weight or less, preferably 20% by weight or less based on the total solid content of the mixture obtained. Use of the solvent in an amount larger than 40% by weight is not desirable because it makes the granulation difficult and requires a longer drying time.

[0031] The granular mixture obtained above may as necessary be dried by a known drying means (e.g. fluidized bed dryer, hot air circulating dryer, vacuum dryer or fluidized bed vacuum dryer) to evaporate the solvent, etc. contained in the mixture.

[0032] The particle diameters of the granular mixture can be 0.03 to 5 mm, preferably 0.1 to 3.0 mm. When the particle diameters are smaller than 0.03 mm, it is difficult to allow the short fiber to be contained in the granular material, which may incur scatter in separator strength. When the particle diameters are larger than 5 mm, no uniform packing density is obtained when the granular mixture is placed in a die for molding and, when a separator of small thickness is produced, molding may become inferior.

[0033] In the present process, next, the granular mixture is made into a fuel cell separator shape. As the method usable for this operation, there can be mentioned, for example, injection molding, injection compression molding and compression molding. Compression molding is preferred.

[0034] In the above step, the molding temperature, the molding pressure and the molding time can be determined depending upon the binder, etc. used. However, there can be employed, for example, a temperature range from room temperature to the curing, melting or vulcanization of the resin, specifically as the molding temperature. The molded material may be subjected to a higher-temperature treatment to chemically stabilize the molded material.

[0035] For improved productivity, it is also possible to produce, in advance, a precursor by a method such as tabbing, extrusion molding, pre-forming, belt press, roll press or the like, and then feed the precursor into a molding machine to obtain a fuel cell separator shape.

[0036] In the process for production of fuel cell separator according to the present invention, it is possible to form, during the molding of the granular mixture into a separator shape, grooves for oxidant gas feeding, grooves for fuel gas feeding, manifold, etc. all necessary for fuel cell separator, in the separator shape. This formation is made possible, for example, by (1) allowing at least either of the upper die and lower die (or core and cavity, or force die and support die, or male plate and female plate) of the die used in compression molding, to have surface unevennesses corresponding to the grooves for oxidant gas feeding, grooves for fuel gas feeding, manifold, etc. of fuel cell separator and (2) using the resulting die for molding of the granular mixture.

[0037] According to the present process, there can be easily obtained a fuel cell separator of small thickness, for example, a fuel cell separator having a thickness of 0.1 to 2.0 mm at the thinnest portion. Moreover, this separator has a deflection in bending of 0.5 mm or more, a bending strength of 4 to 15 kgf/mm², and a bending modulus of elasticity of 2,000 to 6,000 kgf/mm², and has a high strength and high toughness.

[0038] Using the above-mentioned fuel cell separator of the present invention which has a strength necessary for separator even when made in a small thickness, there can be obtained a solid polymer type fuel cell of the present invention (which is compact and highly durable as compared with conventional solid polymer type fuel cells) in the same manner as in conventional solid polymer type fuel cells, that is, by using the present fuel cell separators, a solid electrolyte membranes, gas diffusion electrodes and a sealing members and by clamping the bolts inserted therein, at a given pressure.

5 Description of Preferred Embodiments

10 [0039] The present invention is described in more detail below by way of Examples.

Examples 1 to 25

[0040] A binder, a carbon filler and a carbon fiber all shown in Table 1 or 2 were mixed in a mixer in the amount ratio 15 shown in Table 1 or 2. Into the resulting mixture was dropped methanol in an amount of 15% by weight based on the total solid content of the mixture to produce a granular material having a particle size distribution of 0.1 to 3.0 mm. The granular material was dried in a dryer of 40°C for 3 hours. The dried material was subjected to compression molding at a molding temperature of 150°C at a molding pressure of 150 kg/cm² for 5 minutes to produce a separator for fuel cell 20 having a thickness of 0.3 mm at the thinnest portion. The separator was measured for resistivity, gas permeability, bending strength, bending modulus of elasticity and deflection in bending. The results are shown in Table 1 or 2.

[0041] Incidentally, resistivity was measured by JIS H 0602; gas permeability was measured by the equal pressure method of JIS K 7126; bending strength and bending modulus of elasticity were measured by JIS K 6911; and deflection in bending was taken as the deformation appearing up to the rupture in the test for bending strength and bending modulus of elasticity.

25 [0042] Using each of the separators produced in Examples 1 to 25, together with a solid polymer electrolyte membrane, gas diffusion electrodes and a sealing members, a solid polymer type fuel cell was assembled and operated for 1,000 hours. The cell voltage after 1,000-hour operation was measured as a value when the initial cell voltage was taken as 100. After the 1,000-hour operation, the cell was disintegrated to observe the separator condition. The results are shown in Table 1 or 2.

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Table 1

		Example													
		1	2	3	4	5	6	7	8	9	10	11	12	13	14
Binder	Novolac type phenolic resin	100	100	100	100	100	100	100	100	100	100	100	100	100	100
	Resole type phenolic resin														
Carbon fiber	Scaly graphite (average Particle diameter=30 μm)	290	290	290	290	290	290	290	290	290	290	290	290	290	290
	Average fiber length=0.03mm	69													
	Average fiber length=0.07mm	68													
	Average fiber length=0.1mm	68													
Carbon fiber	Average fiber length=0.4mm	68													
	Average fiber length=0.7mm	68													
	Average fiber length=0.9mm	68													
	Average fiber length=3.0mm	68													
	Average fiber length=6.0mm	68													
Lubricant	Zinc stearate	4.55	4.55	4.55	4.55	4.55	4.55	4.55	4.55	4.55	4.55	4.55	4.55	4.55	4.55
	Resistivity($\text{m}\Omega \cdot \text{cm}$)	60	25	23	25	25	35	60	16	19	19	19	25	31	
	Gas permeability($\text{ml}/(\text{m}^2 \cdot \text{day} \cdot \text{atm})$)	80	50	10	3	3	3	50	80	10	3	3	3	3	50
	Deflection in bending(mm)	0.5	0.5	0.6	0.7	1.0	0.7	0.5	0.5	0.5	0.6	0.7	1.0	0.7	0.5
	Bending strength(kgf/mm ²)	4.2	4.5	6.0	7.0	10.0	7.0	5.5	5.0	5.2	6.0	6.0	7.0	5.0	5.0
	Bending modulus(kgf/mm ²)	5500	4500	4000	3900	3600	4500	5500	5000	4500	3900	3400	4000	5000	
	Cell voltage after 1,000hours(initial cell voltage is taken as 100)	95	95	97	99	99	97	95	95	97	99	99	99	97	
	Separator condition after 1,000-hour operation	No													
	prob-lem	prob-lem	prob-lem	prob-lem	prob-lem	prob-lem	prob-lem	prob-lem	prob-lem	prob-lem	prob-lem	prob-lem	prob-lem	prob-lem	

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Table 2

	Example										
	1.5	1.6	1.7	1.8	1.9	2.0	2.1	2.2	2.3	2.4	2.5
Binder	Novolac type phenolic resin	100	100						100	100	100
	Epoxy resin			100							
	Diallyl phthalate resin				100						
	Polyphenoxy sulfone					100					
	Silicone rubber						100				
Carbon filler	Scaly graphite (average particle diameter=30 μm)	200	800	290	290	290	290	290			145
	Scaly graphite (average particle diameter=80 μm)								290		
	Scaly graphite (average particle diameter=20 μm)									290	
	Acetylene black (average particle diameter=40 μm)										290
	Average fiber length=0.07mm	68	68	10	300	68	68	68	68	68	68
Lubricant	Zinc stearate	4.55	4.55	4.55	4.55	4.55	4.55	4.55	4.55	4.55	4.55
Resistivity($\text{m}\Omega \cdot \text{cm}$)		45	8	23	30	24	26	28	28	30	9
Gas permeability($\text{ml}/(\text{m}^2 \cdot \text{day} \cdot \text{atm})$)		3	45	30	25	3	6	10	20	3	45
Deflection in bending(mm)		0.7	0.5	0.5	0.7	0.7	0.6	0.9	0.5	0.6	0.5
Bending strength(kgf/mm ²)		5.0	4.2	4.0	6.0	6.8	5.5	6.0	4.5	6.0	4.3
Bending modulus(kgf/mm ²)		4500	4000	4000	3500	3600	4500	3500	4500	3900	3400
Ceil voltage after 1,000 hours (initial cell voltage is taken as 100)		97	95	95	95	99	98	98	95	97	95
Separator condition after 1,000-hour operation	No problem	No problem	No problem	No problem	No problem	No problem	No problem	No problem	No problem	No problem	No problem

Examples 26 to 28

[0043] A separator for fuel cell was produced in the same manner as in Example 5 except that the granular mixture used had a particle size distribution shown in Table 3. The separator was measured for resistivity, gas permeability, bending strength, bending modulus of elasticity and deflection in bending in the same manners as in Examples 1 to 25. The results are shown in Table 4. A polymer type fuel cell was assembled in the same manner as in Examples 1 to 25, and measured for cell voltage and separator condition in the same manners as in Examples 1 to 25. The results are shown in Table 4.

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Table 3

Particle size distribution	0.03-0.1 mm	0.1-3.0 mm	3.0-5.0 mm
Example 26	80%	20%	0%
Example 27	25%	50%	25%
Example 28	0%	20%	80%

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Example 29

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[0044] A separator for fuel cell was produced in the same manner as in Example 5 except that injection molding was used. The separator was measured for resistivity, gas permeability, bending strength, bending modulus of elasticity and deflection in bending in the same manners as in Examples 1 to 25. The results are shown in Table 4. A polymer type fuel cell was assembled in the same manner as in Examples 1 to 25, and measured for cell voltage and separator condition in the same manners as in Examples 1 to 25. The results are shown in Table 4.

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Example 30

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[0045] A separator for fuel cell was produced in the same manner as in Example 5 except that compression injection molding was used. The separator was measured for resistivity, gas permeability, bending strength, bending modulus of elasticity and deflection in bending in the same manners as in Examples 1 to 25. The results are shown in Table 4. A polymer type fuel cell was assembled in the same manner as in Examples 1 to 25, and measured for cell voltage and separator condition in the same manners as in Examples 1 to 25. The results are shown in Table 4.

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Table 4

		Example				
		26	27	28	29	30
Binder	Novolac type phenolic resin	100	100	100	100	100
Carbon filler	Scaly graphite (average particle diameter=30 µm)	290	290	290	290	290
Carbon fiber	Average fiber length=0.07mm	68	68	68	68	68
Lubricant	Zinc stearate	4.55	4.55	4.55	4.55	4.55
Resistivity(mΩ · cm)		60	58	60	120	100
Gas permeability(ml/m² · day · atm)		80	75	80	110	100
Deflection in bending(mm)		0.5	0.5	0.5	0.5	0.5
Bending strength(kgf/mm²)		4.2	4.4	4.2	4.0	4.0

Table 4 (continued)

	Example				
	26	27	28	29	30
5	Bending modulus(kgf/mm ²)	5500	5000	5500	5000
10	Cell voltage after 1,000 hours(initial cell voltage is taken as 100)	95	95	95	93
15	Separator condition after 1,000-hour operation	No problem	No problem	No problem	No problem

Comparative Example 1

[0046] A separator for fuel cell was produced in the same manner as in Example 1 except that no carbon fiber was used and the amount of carbon filler was increased to 358 parts by weight. The separator was measured for resistivity, gas permeability, bending strength, bending modulus of elasticity and deflection in bending in the same manners as in Example 1. The results are shown in Table 5. A polymer type fuel cell was assembled in the same manner as in Example 1, and measured for cell voltage and separator condition in the same manners as in Example 1. The results are shown in Table 5.

Comparative Example 2

[0047] A separator for fuel cell was produced in the same manner as in Example 5 except that no carbon filler was used and the amount of carbon fiber was increased to 358 parts by weight. The separator was measured for resistivity, gas permeability, bending strength, bending modulus of elasticity and deflection in bending in the same manners as in Example 1. The results are shown in Table 5. A polymer type fuel cell was assembled in the same manner as in Example 1, and measured for cell voltage and separator condition in the same manners as in Example 1. The results are shown in Table 5.

Comparative Example 3

[0048] A separator for fuel cell was produced in the same manner as in Example 8 except that no carbon filler was used and the amount of carbon fiber was increased to 358 parts by weight. The separator was measured for resistivity, gas permeability, bending strength, bending modulus of elasticity and deflection in bending in the same manners as in Example 8. The results are shown in Table 5. A polymer type fuel cell was assembled in the same manner as in Example 8, and measured for cell voltage and separator condition in the same manners as in Example 8. The results are shown in Table 5.

Comparative Examples 4 to 9

[0049] A binder, a carbon filler and a carbon fiber all shown in Table 5 were mixed in a mixer in the amount ratio shown in Table 5. Into the resulting mixture was dropped methanol in an amount of 15% by weight based on the total solid content of the mixture to produce a granular material. The granular material was dried in a dryer of 40°C for 3 hours. The dried material was subjected to compression molding at a molding temperature of 150°C at a molding pressure of 150 kg/cm² for 5 minutes to produce a separator for fuel cell having a thickness of 0.3 mm at the thinnest portion. The separator was measured for resistivity, gas permeability, bending strength, bending modulus of elasticity and deflection in bending, in the same manners as in Examples. The results are shown in Table 5. Using each of the separators produced above, together with a solid polymer electrolyte membrane, gas diffusion electrodes and a sealing members, a solid polymer type fuel cell was assembled and operated for 1,000 hours. The cell voltage after 1,000-hour operation was measured as a value when the initial cell voltage was taken as 100. After the 1,000-hour operation, the cell was disintegrated to observe the separator condition. The results are shown in Table 5.

Table 5

		Comparative Example								
		1	2	3	4	5	6	7	8	9
Binder	Novolac type phenolic resin	100	100	100	100	100	100	100	100	100
Carbon filler	Scaly graphite (average particle diameter 30 μm)	358			100	1000	290	290	290	290
Carbon fiber	Average fiber length=0.07mm	358		68	68	5	600			
	Average fiber length=0.01mm							68		
	Average fiber length=6mm		358							
	Average fiber length=12mm									68
Lubricant	Zinc stearate	4.55	4.55	4.55	4.55	4.55	4.55	4.55	4.55	4.55
	Resistivity(m Ω cm)	20	80	400	1000	400	26	500	28	23
	Gas permeability(ml/m \cdot day·atm)	100000	100000	1000	5000	10000	20000	100000	100000	100000
	Deflection in bending(mm)	0.2	0.3	0.2	0.2	0.2	0.2	0.2	0.2	0.2
	Bending strength(kgf/mm 2)	2.1	1.5	7.0	3.0	2.0	2.0	1.9	2.0	
	Bending modulus(kgf/mm 2)	5500	4500	3000	3500	3600	4500	3500	4500	4500
	Cell voltage after 1,000 hours (Initial cell voltage is taken as 100)	Note(2)	Note(1)	Note(1)	25	Note(2)	Note(2)	Note(2)	Note(2)	
	Separator condition after 1,000-hour operation	Cracked	Cracked	No problem	No problem	Crazed	Cracked	Cracked	Cracked	

Note(1): Voltage effect was too large owing to high resistance, and power generation was impossible.

Note(2): Gas leakage was too striking, and power generation was impossible.

[0050] As is clear from the above Examples and Comparative Examples, the separator for fuel cell according to the present invention has a strength necessary for separator even when made in a small thickness, generates no crack at the time of fuel cell assembling or during the power generation of assembled fuel cell, and has high reliability.

[0051] The process for production of fuel cell separator according to the present invention can produce an excellent fuel cell separator such as mentioned above, without requiring any graphitization or machining step.

[0052] The solid polymer type fuel cell of the present invention using the above separator has very high durability.

5 Claims

1. A separator for fuel cell, formed using a base material obtained from a composition comprising at least a binder, a powdery carbon filler having an average particle diameter of 10 nm to 100 μm , and a short fiber having an average fiber length of 0.03 to 6 mm, in which composition the amount ratio of the above three components is such that the amount of the powdery carbon filler is 200 to 800 parts by weight and the amount of the short fiber is 10 to 300 parts by weight, both per 100 parts by weight of the binder.
2. A separator according to Claim 1, wherein the short fiber is at least one kind of fiber selected from a carbon fiber, a pyrolyzed carbon fiber and a stainless steel fiber.
3. A process for producing a separator for fuel cell, which comprises mixing at least a binder, a powdery carbon filler having an average particle diameter of 10 nm to 100 μm , and a short fiber having an average fiber length of 0.03 to 6 mm in such an amount ratio that the amount of the powdery carbon filler becomes 200 to 800 parts by weight and the amount of the short fiber becomes 10 to 300 parts by weight, both per 100 parts by weight of the binder, granulating the resulting mixture into a granular material of 0.03 to 5 mm in particle diameter, and molding the granular material into a separator shape.
4. A process according to Claim 3, wherein the granulation of the mixture is conducted using a solvent.
- 25 5. A solid polymer type fuel cell using a separator set forth in Claim 1 or 2.

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